Characterization of Material Properties at Terahertz Frequencies

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Introduction

In the realm of materials science, terahertz frequency measurement systems provide significant utility in the characterization of material properties. With support primarily from the U.S. Army National Ground Intelligence Center (NGIC), Submillimeter Technology Laboratory (STL) researchers have been advancing the field of terahertz technology to the application of modeling millimeter-wave and microwave radar for more than a decade.[(1)] Research in modeling radar requires design of a wide range of measurements systems using current submillimeter-wave source/detector technology, establishment of precise calibration standards, production of high-fidelity scale replicas of complex metallic structures, and scaling of millimeter-wave dielectric properties of composites at submillimeter-wave frequencies. This paper explores four measurement techniques typically employed by STL to perform the characterization of materials:

(1) laser-based submillimeter-wave ellipsometry;
(2) high-precision reflectometry;
(3) laser-based Brewster's angle measurements;
and, (4) FIR Fourier transform spectroscopy (FTS).

While trade-offs exist between precision and ease of implementation, each technique provides unique capabilities. As the most precise method of measuring a material's submillimeter-wave optical properties, laser-based ellipsometry can measure the extinction coefficient to a precision of a few percent and has the advantage of being able to evaluate opaque as well as transparent materials. High-precision submillimeter-wave reflectometry allows reflectivity measurements to an uncertainty of 0.1%. Laser-based Brewster's angle measurements, while not highly precise, allow rapid determination of dielectric constants for lossy materials based solely on front-surface reflectivity; and FIR Fourier transform spectroscopy (FTS) allows one to characterize the optical properties of materials as a function of frequency.

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Laser-Based Submillimeter-Wave Ellipsometry

Ellipsometry determines material optical properties by measuring a change in the reflected polarization state from the sample. By knowing the incident polarization state and measuring the reflected polarization state as a function of several incident angles, it is possible to derive a material's refractive index (n) and extinction coefficient (k).(2) Using a mathematical representation developed by D.A. Holmes(3), the STL research team designed six quartz quarter-wave retardation plates (QWPs) to analyze the reflected polarization state at six specific frequencies covering 500 GHz to 2.5 THz.

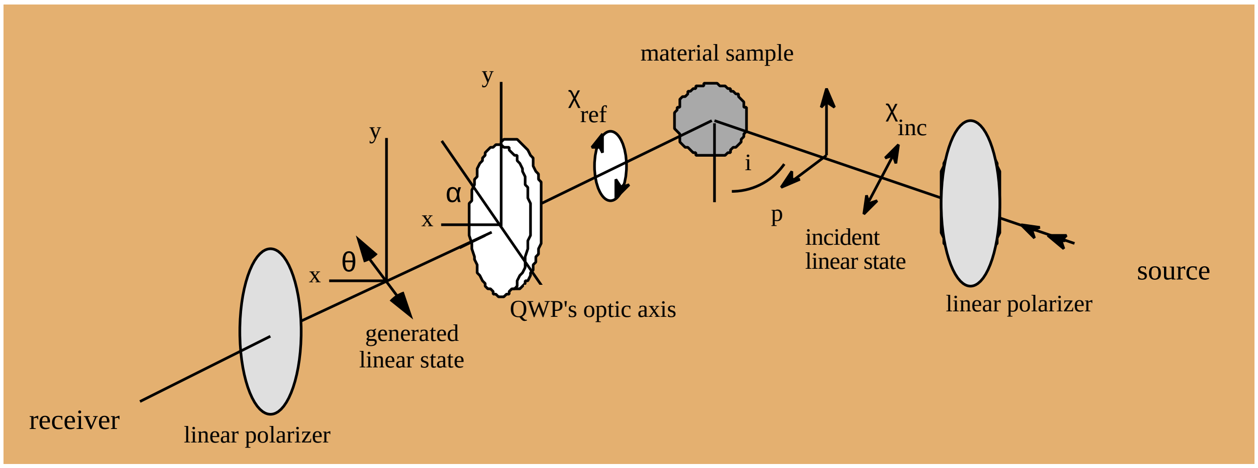


Figure 1. The ellipsometric measurement technique using a quarter-wave plate.

Shown schematically in Figure 1, the submillimeter-wave ellipsometry technique implemented by STL(4) relies on a QWP analyzer whose incident angle is adjusted until its relative phase shift is $\pi/2$. As originally suggested by Oldham(5), the tilted QWP analyzer can then transform the sample's reflected radiation from some unknown elliptical state, χ_{ref}, to a linear state. The orientation of the linear state is made known via the high-speed rotating linear polarizer. The data on the angular orientation of the QWP and the linear state are reduced via equations 1, and the reflected polarization state from the material is inferred.

$$\chi_{ref} = \frac{E_s}{E_p} = f(\alpha, \theta, t_r) \quad \text{where} \quad |\chi_{ref}| = \frac{1 + t_r^2 \tan^2\alpha \tan^2(\alpha+\theta)}{\tan^2\alpha + t_r^2 \tan^2(\alpha+\theta)} \qquad [1]$$

$$\text{and} \quad \varphi_{ref} = \varphi_p - \varphi_s = -t_r(\cot\alpha + \tan\alpha)\tan(\alpha+\theta)$$

The reflected state also depends upon the transmissivity ratio of the ordinary (s) and extraordinary (p) components in the birefringent QWP. Taking this ratio and using the fundamental equation of ellipsometry(2), equa. 2, one can determine the amplitude and phase ratio between the s and p components. The automated system is shown in Figure 2. Since the radiation is modulated via the

rotating linear polarizer, a two-pase lock-in amplifier is used to demodulate the signal to determine the orientation of the linearly polarized state created between the QWP and the linear polarizer.

$$\frac{\chi_{inc}}{\chi_{ref}} = \frac{E_{s_i}}{E_{p_i}} \times \frac{E_{p_r}}{E_{s_r}} = \frac{R_p}{R_s} = \rho = \tan\psi\, e^{i\Delta} \qquad [2]$$

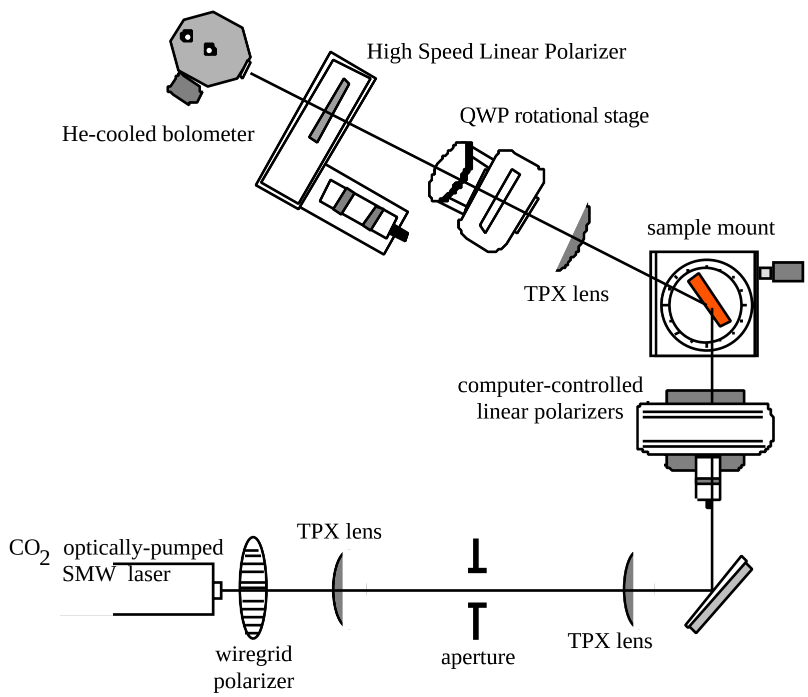


Figure 2. STL’s automated submillimeter-wave ellipsometric measurement system.

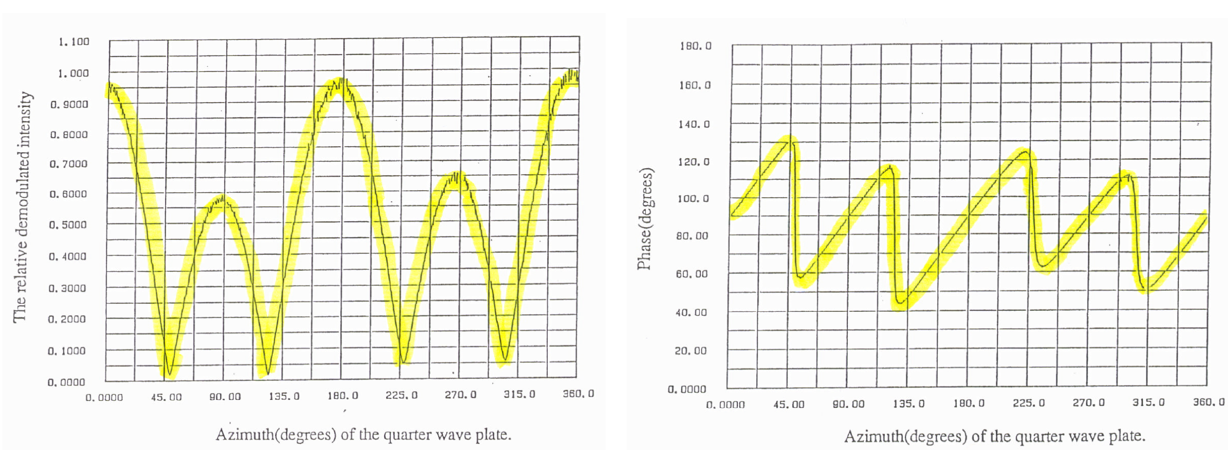


Figure 3. The demodulated intensity as a function of the QWP’s azimuth.

Depicted in Figure 3 is calibration data from the ellipsometer, obtained using vertically polarized radiation. On the left is the demodulated intensity as a function of rotation of the QWP through

360°. The peaks with higher and lower amplitudes correspond to the ordinary and extra-ordinary axes of the QWP. The ratio between peaks is the transmissivity ratio for the QWP (t_r), which is dependent on the birefringence of the quartz QWP. By determining the orientation of the QWP for each peak (α) and measuring the phase using the two-phase lock-in amplifier (θ, see right hand graph of figure 3), the unknown reflected polarization state from the sample can be calculated, equation 1.

As exemplified by figure 4, STL's A.J. Gatesman was able to establish the submillimeter-wave optical properties of high-purity silicon by obtaining experimental data for a sample over a range of incident angles.(6) The refractive index and extinction coefficient of a material is determined by using them as variables in the Fresnel equations, and finding the best fit between experimental data and theoretical prediction (the continuous line in Figure 4). As shown in Table 1, the optical properties are obtained with very high precision and accuracy with the extinction and absorption coefficients given for a one cm sample thickness. Maximum uncertainties are $\Delta n \approx \pm 0.002$ and $\Delta k \approx \pm 0.0001$, however, the thickness and incident angle of the plate must be known to within ±1 µm and ±0.5°, respectively.

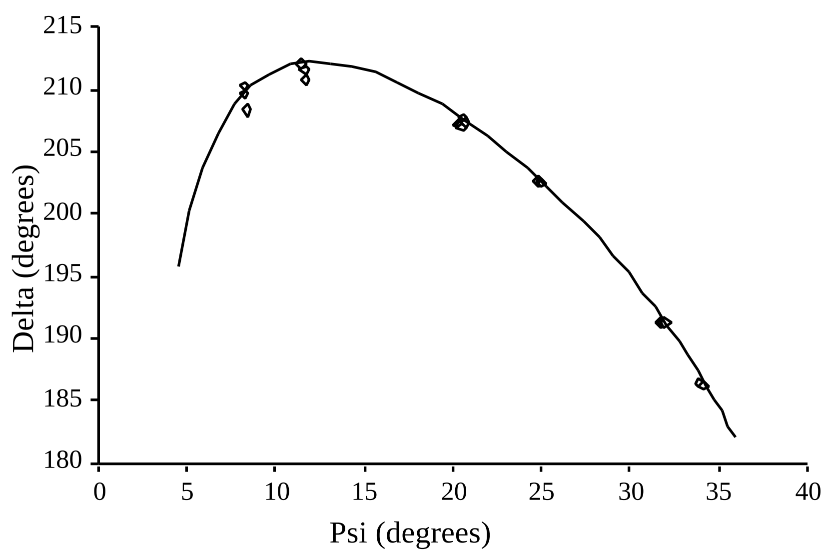


Figure 4. Ellipsometric measurement of high purity silicon at a wavelength of 513.0157 µm.

Since the ellipsometric measurement technique relies only on the relative amplitudes between the reflected s and p components and not the absolute reflectivity of the sample material, the variation in intensity of the incident laser radiation does not cause loss of accuracy. Silicon etalons could then be established as calibration reflection standards, based on the small uncertainties achieved in measuring their optical properties and by exploiting the low loss nature of the material.(7,8)

Table 1. The SMW Ellipsometrically measured Optical Properties of Silicon

λ (μm)	**ν (THz)**	**refractive index**	**extinction coeff**	**absorption coeff**
117.72748	2.55	3.4162±0.0002	0.00007 ±0.00002	0.07±0.02
191.84803	1.56	3.4160±0.0002	0.00006 ±0.00003	0.04±0.02
236.6008	1.27	3.4164±0.0002	0.00007 ±0.00004	0.04±0.02
513.0157	0.58	3.4164±0.0002	$0.00004^{+0.00008}_{-0.00004}$	$0.01^{+0.02}_{-0.01}$

High-Precision Reflectometry

A.J. Gatesman used the silicon etalons as reflection standards when developing his high-precision submillimeter-wave reflectometer.(6,9) As STL's second method of characterizing materials, reflectometry was used to evaluate the high frequency performance of metals.(9) A block diagram of the optics and measurement components of the reflectometer are shown in Figure 5. To minimize standing wave problems as well as feedback to the laser, attenuators are inserted in the beam since the system's signal-to-noise ratio is sufficient to allow this. In staring mode, this system achieves reflectivity uncertainties on the order of ±0.1%.

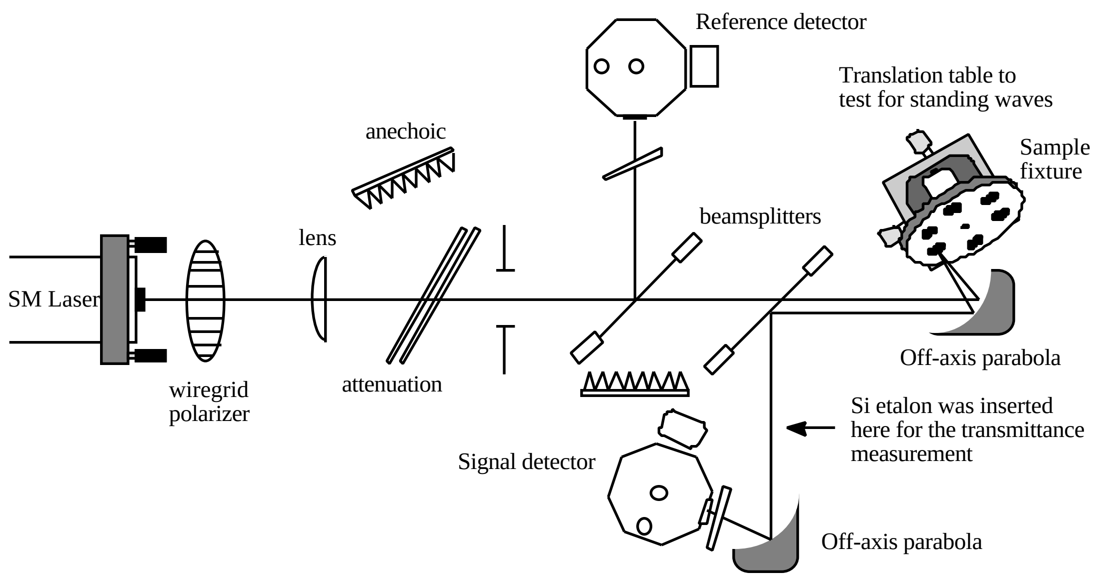


Figure 5. STL's Submillimeter-Wave Reflectometer.

The largest difficulty in performing precise reflection measurements is maintaining alignment to the incident beam when samples and standards are interchanged. The sample fixture is designed to

have spring-loaded samples and standards mounted against a precision ground flat surface to minimize alignment uncertainties. The sample stage is rotated with a 4 inch diameter air bearing to obtain interchangeability on the order of ±0.03°. As shown in Table 2, STL's submillimeter-wave reflectometer allows one to measure the differences in reflectivity of metals with ±0.1% uncertainty.

Table 2. The Submillimeter-Wave Reflectivity of Metals

Metal	**R (0.58 THz)**	**R (2.55 THz)**
Copper	0.997	*
Silver	0.996	0.995
Gold	0.994	0.994
Aluminum	0.995	0.994
Nickel	0.994	0.991
Chromium	0.993	0.974

Laser-Based Brewster's Angle Measurement Technique

For rapid determination of the submillimeter-wave optical properties of lossy materials, STL's researchers implemented a third measurement method which exploits the relationship between Brewster's angle, θ_B, and a material's refractive index (i.e. $n = \tan \theta_B$).[10] Shown in Figure 6, this optical arrangement measures a material's reflectivity from 0° to 85° incidence, monostatically. The transmitter and receiver remain fixed while rotating the dihedral sample mount. One face of the dihedral is a gold first surface mirror while the second surface is a vacuum chuck for quickly securing a sample.

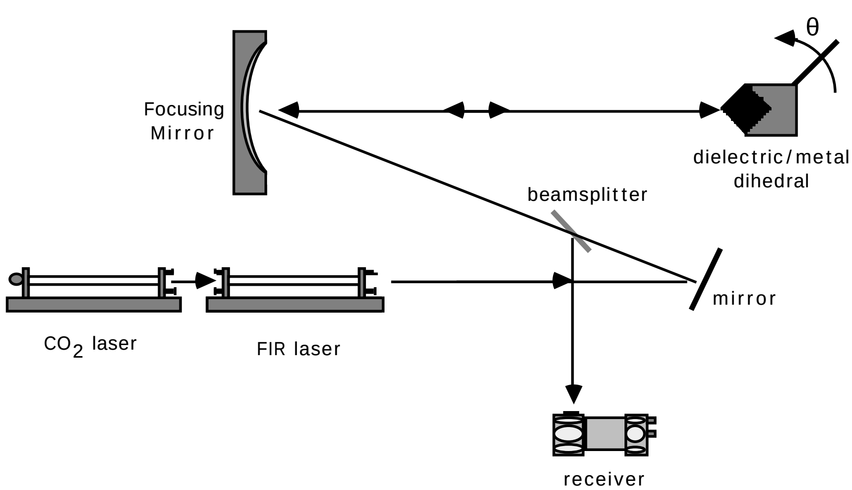
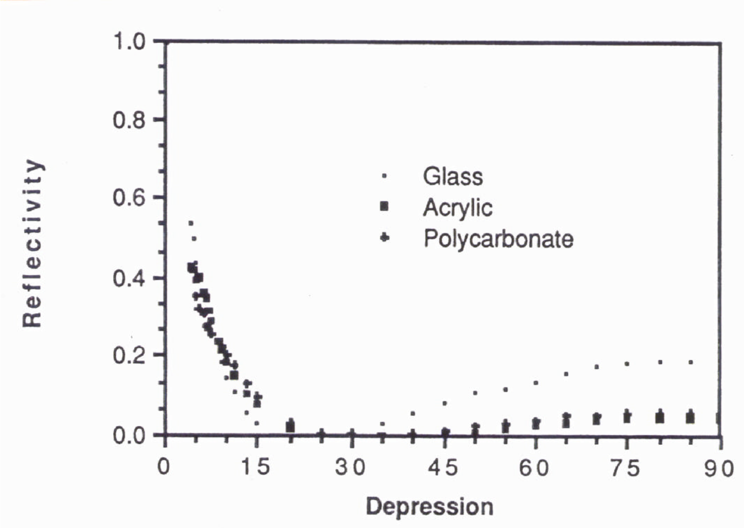


Figure 6. STL's Laser-Based Brewster's Angle Measurement System



Material	Refractive Index (n)
acrylic	1.43±0.02
polycarbonate	1.60±0.02
glass	2.62±0.02

Figure 7. The measured reflectivity as a function of incident angle for three materials at 1.5 THz.

The measured reflectivity data provides Brewster's angle, θ_B, to a precision of ±0.1°. While the accuracy of this system is no where near comparable to the two previously discussed systems, its optical configuration is simple to setup and measurements can be made rapidly. The measured reflectivity of glass, acrylic, and polycarbonate are displayed in Figure 7. To evaluate the refractive index values given below, a curve fitting technique using the Fresnel reflection coefficient (p-wave) was applied to the reflectivity measurements. This technique is a relatively insensitive measure of a material's extinction coefficient, k.

Far-Infrared Fourier Transform Spectroscopy

As the fourth technique for measuring optical properties of materials, far-infrared Fourier transform spectroscopy (FIR FTS) enables one to characterize materials as a function of frequency.(11,12) As shown in figure 8, transmission (and/or reflection) measurements are performed for a sample over the submillimeter wavelength region and the frequency dependent complex refractive index is ascertained using the Fresnel coefficient multiple reflection theory of etalons. Ultimately, determination of the refractive index, n, to an uncertainty of ±0.01 is possible by either knowing the order of a fringe, m, using the expression: $n = m/(2\ t\ \nu)$, or by the difference between the orders of two fringes, m_1 and m_2,

where $2\,t\,n\,\nu_1 = m_1$ and $2\,t\,n\,\nu_2 = m_2$

therefore $$n = \Delta m/(2\,t\,\Delta\nu). \quad [3]$$

The uncertainty in determining a material's refractive index by this technique is a function of the sample thickness, t, as well as the material's index of refraction.

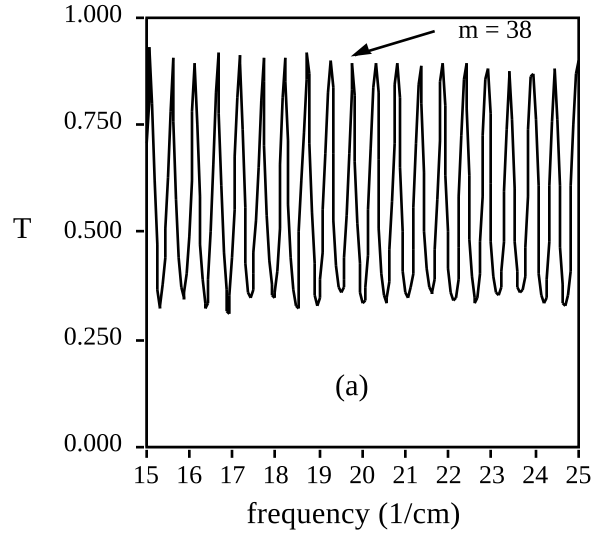


Figure 8. Locating a transmission peak in far-infrared Fourier transform spectroscopy data.

Applications

STL's interest in materials research revolves about an effort in tailoring the dielectric properties of composite structures, or finding materials with optical properties at THz frequencies that simulate materials at millimeter-wave or microwave frequencies. Based on measurements using the four characterization techniques, STL has developed a variety of artificial dielectric materials for bulk and thin film applications, and have tailored their optical properties for the fabrication of frequency-selective absorbing structures.[13] A variety of binders such as vinyl acetate, silicone, polyethylene, and epoxy resins have been loaded with powdered agents such as carbon, silicon, and stainless steel flake, in order to achieve the desired submillimeter–wave optical properties.

The optical properties of these powder loaded binders proved ideal for creating an anechoic layer on metal surfaces. As shown in Figure 9, a thin film provides the vehicle for which phase and amplitude matching of the incident electric field can occur. Calculation of the structure's optical behavior can be performed using the Fresnel equations. With a reflectivity of ≈ 1 for aluminum, the resonant structure's theoretical reflectivity is approximated as

$$R \sim \left| \frac{r + e^{-2i\beta}}{1 + r\, e^{-2i\beta}} \right| \tag{4}$$

where $r = \frac{N-1}{N+1}$ and $\beta = 2\pi t\, N/\lambda$

are the thin film's front surface reflectivity and phase thickness, respectively.

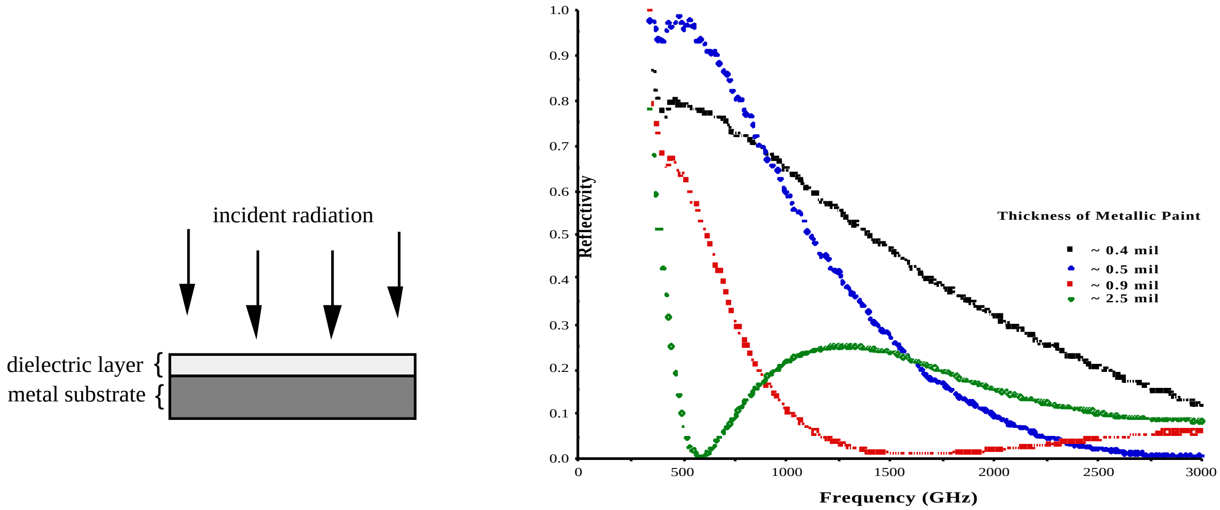


Figure 9. The reflectivity of four Dällenbach layers with a constant loading ratio.

As evident by equation 4, a phase thickness of a quarter wavelength will cause destructive interference. If the amplitude of the electric field reflected from the quarter-wavelength thin film's front surface equals that of the back surface which suffers absorption, all the incident electric field is reflected back into the material and complete resonant absorption is established. A reflectivity null at the frequency of choice can be achieved by the proper choice of materials. STL's realization of the resonant structure using a vinyl acetate film loaded with stainless steel flake is illustrated in Figure 10.

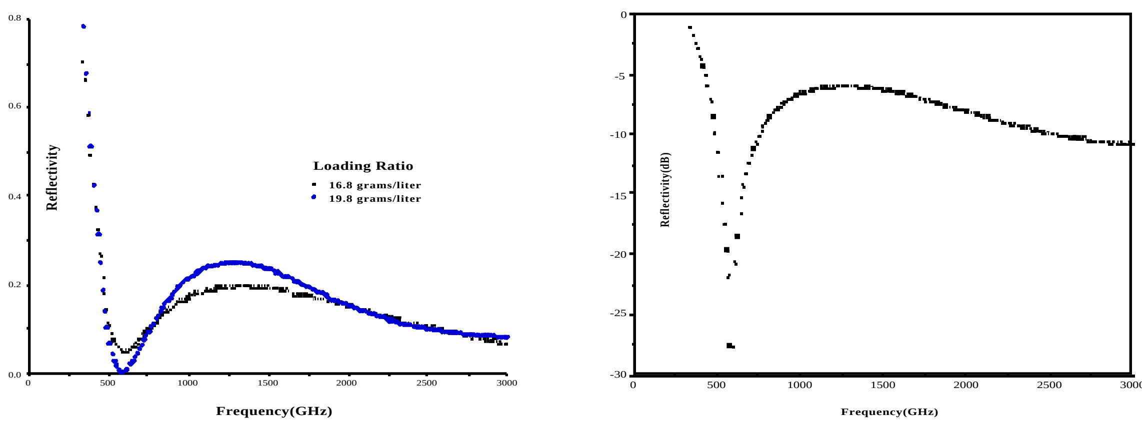


Figure 10. The reflectivity of a Dällenbach layer optimized for performance at 0.584 THz.

Summary

In summary, the Submillimeter Technology Laboratory research staff has developed a complement of precision measurement systems specifically for evaluating the optical properties of materials at

THz frequencies. STL has established the calibration standards for performing reflectivity measurements to a precision of ±0.1%, developed a variety of artificial dielectric materials for bulk and thin film applications, and tailored their optical properties for the fabrication of frequency selective absorbing structures. All these techniques should prove eminently useful for further investigations of materials properties in the terahertz frequency regime.

Acknowledgements

Several colleagues at the University of Massachusetts Lowell's Submillimeter Technology Laboratory have made significant contributions to this work. In the early '80s, Dr. Jerry Waldman, while working at MIT Lincoln Laboratory, developed techniques for scaling millimeter-wave radar at submillimeter-wave frequencies. He moved the laboratory to Lowell in 1982 and developed a research group for designing measurement systems to acquire scaled radar data at terahertz frequencies. As the performance of these measurement systems improved, the fidelity of scaled replicas became a driving issue along with modeling the dielectric properties of composite materials. By the direction of W.E. Nixon, support to STL from the U.S. Army National Ground Intelligence Center diversified to encompass a wide range of materials research. Under support for this program the author received his Ph.D. for the design and fabrication of a submillimeter-wave ellipsometer and A.J. Gatesman received a Ph.D. for developing the high-precision reflectometer.

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